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NATURAL RADON EXHALATION RATE FROM FERTILIZER USED IN BASRA GOVERNORATE/IRAQ

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ABSTRACT

A passive method to measure the radon activity concentration in fertilizer samples, by the Solid State Nuclear Track Detectors (SSNTDs) of type CR39 has been used in the present work, also radon exhalation rate and effective radium content. The RAD7 radon detector has also been used as an active method for measuring the radon activity concentrations and comparing it with the CR39 results. The results reveal that the highest radon concentration, area exhalation rate, mass exhalation rate and effective radium content of 1197±97 Bq/m^3, 2408 mBq/m^2.h, 51.22 mBq/Kg.h and 6.782 Bq/kg respectively, were found in Humi- plan fertilizer; while the lowest values of these parameters were found to be 13±4 Bq/m^3, 26.8 mBq/m^2.h, 0.34 mBq/kg. h and 0.046 Bq/kg respectively in potassium sulphate fertilizer. It is concluded that the radon exhalation rate from the fertilizers do not significantly contribute to the natural environmental radioactivity.

KEYWORDS: Fertilizers Samples, RAD7, SSNTDs, Radon, Exhalation Rates, Effective Radium

INTRODUCTION

The fertilizer play a prime role in agriculture and raw material used in production of some fertilizers is phosphate ore containing various amounts of natural radioactive elements like ^{226}Ra. Most of the phosphate fertilizers are considered as waste and is stockpiled or discharged into the environment [1]. The effect of Phosphate disposal into environment is the possible increases in radio- nuclides in soils or in groundwater and consequential ingestion by humans through exposure routes such as drinking water and food chain [2]. The natural radioactivity in fertilizer may vary considerably from one type to another depending on the chemical structure of fertilizer and normally fertilizers used for agriculture in Iraqi planet are rich in phosphate ores [3]. The phosphate ores, specially the sedimentary ores, specially the disposal ores, can be significantly enriched with naturally uranium (^{235}U, ^{238}U) and their daughters [4]. There is a direct relationship between uranium and P_2O_5 content in the fertilizer and then several studies have been noted that the concentration of uranium follows the concentration of P_2O_5 in various fertilizers [5-6]. In NPK fertilizer, potassium component augment the natural radioactivity because of the presence of radioactive ^{40}K, whose natural abundance in potassium ore is 0.0018%. Since the starting materials of the fertilizers is phosphate rock used in wet process by attack of sulphuric acid in fertilizer industry contain varying amount of uranium. The ^{238}U remains concentrated into phosphoric acid while ^{226}Ra, ^{210}Po, ^{232}Th and ^{210}Pb precipitated out as sulphate salt concentrated in phosphogypsum as the byproduct. The uranium either in form of $[U(SO_4)_2]$ or $[UO_2(SO_4)]$ in phosphoric acid is water soluble, remains in phosphoric acid which is used for the fertilizers production, thus the uranium content of fertilizers is expected to be high [7-9]. The environmental impact of fertilizer production depends on the raw materials, production processes and the status of the pollution control equipment. In addition, fertilizer plants cause environmental harm through emissions of process specific chemicals into the air,

discharges into water, and storage and solid waste problems. Actions should be undertaken to minimize the missions and to clean up spills and solid wastes after using the fertilizers [10]. Globally, the revelation that physical processing of phosphate ore does not alter the radionuclide concentration during the operational processes in fertilizer plants led to legislations concerning discharges in gaseous, liquid and solid forms. Despite public concern about radiological impact of phosphate fertilizer on its immediate environment, there has not been any study, to the best of the authors' knowledge, in Basra governorate. This study carried out near an active phosphate fertilizer plant in Basra to assess the potential radiological impact of phosphate fertilizer plant on its immediate environment. The presence of radium and hence emission of the radon gas in the fertilizer is expected, has a high potential for causing biological damage through continuous irradiation of human skeleton over many years and may induce bone cancers[11]. Emanation of radon gas into air occurs as a product of uranium (^{238}U) and thorium (^{232}Th) decay chains, respectively. The short-lived decay products of radon are responsible for most of the hazards by inhalation. The hazard of radon comes from its radioactive progeny, which uses their physical properties to spread or attach like aerosols do, trapped in the lung and depositing their alpha-particle energies in the tissue, producing higher ionization density than beta particles or gamma-rays. Lung cancer, skin cancer, and kidney diseases are the health effects attributed to inhalation of radon-decay products [12-14]. The sources of radon gas are the building materials and its components, ground water, and soil. The radiological impact from the above nuclides is due to radiation exposure of the body by the gamma rays and irradiation of the lung tissues from inhalation of Radon and its progeny. From the natural risk point of view, it is necessary to know the dose limits of public exposures and to measure the natural environmental radiation level provided by ground, air, water, foods, building interiors, etc, for the estimation of the exposures to natural radiation sources. [15-17].

The aim of the present study is to measure the radon gas concentration, exhalation rates and effective radium mass from fertilizer using passive, SSNTDs method, and active method, using RAD7 instrument, to provide useful information in the monitoring of environmental contamination by natural radioactivity, and this is due to the lack of studies concerning the assessment of radium contents and radon exhalation rates of materials having daily live usage in Basra governorate.

MATERIALS AND METHODS

Fertilizer Samples

Fertilizers are usually used in reclaiming the land and improving the properties of crops. Many types of fertilizers used in Basra governorate like Super Phosphate, Urea Sulfate, NPK, and Ammonium Nitrate Sulfate. Sixteen fertilizers samples were available in the local market in Basra Governorate and the regional fertilizer factory.

Passive Technique

The fertilizer samples were dried in the oven at 110 ^{o}C for 24h, milled in grinder, and sieved. Each dried sample of fertilizer placed in the bottom (5 cm height) of closed hard plastic cylinder container of 30 cm height and diameter 7cm and covered by 5mm sponge, as shown in Figure 1[18]. The 1.5x1.5cm^2 SSNTDs films (CR39) placed on the bottom of the cover of the containers, to allow irradiation process with radon only to take place. After 90 days of irradiation the detectors was removed carefully from the cylinder and etched with NaOH solution with condition 6.25N at 70 ^{o}C for 7 hour. Then the detectors are washed many times by distilled water and dried with tissue papers. The numbers of tracks due to alpha particles interaction are counted by the means of optical microscope 400X.

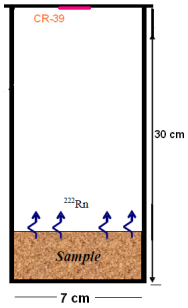

Figure 1: The Hard Plastic Cylinder Used as Radon Irradiator

Radon gas concentration is given by; [19]

$$A_{Rn} = \frac{\rho}{tK} \quad (1)$$

Where ρ is track density in Tr/cm^2, t exposure time in day and K the calibration factor in Tr/cm^2.day / Bq.m^{-3}. The value of K will depend on the height and radius of the measuring can.

At the equilibrium state, final activity of radon exhalation from each sample inside the can is given by [20-21]

$$E_{ex} = \frac{AV\lambda/S}{T+(e^{-\lambda T}-1)/\lambda} \quad (2)$$

Where E_x is exhalation rate in unit Bq m^{-2}.d^{-1}, A is integrated exposure measured by the detector in unit Bq m^{-3} d, λ is radon decay constant, T, the exposure time, V the volume of the can anSd S is the surface area covered by the can.

The radon exhalation rate in terms of mass is calculated from the relation;

$$E_M = \frac{AV\lambda/M}{T+(e^{-\lambda T}-1)/\lambda} \quad (3)$$

Where E_M expressed in Bq kg^{-1}h^{-1} and M is the mass of the sample.

The effective radium content in the sample could be calculated from;

$$A_{Ra} = \frac{\rho hS}{KMt} \quad (4)$$

where ρ is track density recorder, h distance between the detector and sample, S surface area of sample and t exposure time.

Active Technique

A radon gas analyzer RAD7 instrument (DURRIDGE Company USA) was used to measure radon emanation from the ore samples. The ore sample was loaded into 1 l used as an emanation cylindrical container, shown in figure 2. The high of the container 25 cm, to insure radon detection only, was used as the radon accumulation chamber. The alpha RAD7 detector was operated in grab mode for 2days protocol, with cycle 1h and recycle 48. The removable lid was equipped with two gas-tight tubes, one used to pump radon gas in RAD7 chamber (ZnS) and the other used to pump fresh air to the container. The system is a closed loop in which the gas circulates continuously. The experiment was performed at a relative humidity less than 10%, 18-25 ^{o}C and normal room atmospheric pressure. After the sample is place in the accumulation chamber and left for two days for equilibrium, before measurement. The chamber connected to the

instrument as shown in figure 2.The filtered air decays inside the monitor chamber, producing detectable alpha emitting progeny, particularly the polonium isotopes. A high voltage of 2500 V is applied to the chamber walls. The solid-state silicon detector converts alpha radiation directly to an electrical signal discriminating the electrical pulses generated by α-particles from the polonium isotopes (^{218}Po, ^{216}Po, ^{214}Po, ^{212}Po) with energies of 6.0, 6.7, 7.7 and 8.8 MeV, respectively. Using this approach, it is possible to use only the ^{218}Po peak for ^{222}Rn and ^{216}Po for ^{220}Rn, obtaining a rapid equilibrium between polonium and radon nuclides, because the equilibrium between ^{218}Po and ^{222}Rn is achieved in about 15 min (about five times the half-life of ^{218}Po), and between ^{216}Po and ^{220}Rn in a few seconds. The ^{222}Rn growth curve monitored with cycle times from 15 minutes up to 2 hours for a day in order to calculate the exhalation rate that is proportional to the slope of the growth curve [22]. The concentration of radon emanated from each fertilizer sample inside the emanation container was allowed to build up with time and it was measured in 1h cycle for an average time of 48h.The build-up radon activity inside the emanation container follows the equation

$$A_t = A_o\left(1 - e^{-\lambda t}\right) \tag{5}$$

where λ is the decay constant of the nuclide concerned and A_o is the final value of the activity at $t \approx 7.2T_{1/2}$, which approximately 27.5 days for radon. To translate the number of counted tracks into radon concentrations, a calibration made by using a combination of both active and passive methods.

The Calibration Factor

Calibration factors are the quantities, which are used for converting the observed track density on SSNTD after etching to the activity concentrations of the species of interest. If ρ denotes the track densities in (Track/cm^2.d) observed on a SSNTD due to exposure in a given mode to a concentration C in Bq m^{-3} of given species for a time t, it is obvious that one use equation (1) to calculate this parameter. The experimental setting used, shown in figure 2, is a hybrid between active and passive methods. We used a cylindrical tube (7cemx30cm) as a radon chamber; the radon source was ^{226}Ra with activity 48.1 kBq, was left for four weeks in the bottom of closed chamber to reach saturation. After this time, the detectors (CR39) are exposed to radon for different time (2, 3, 4, 7 days). The activity concentrations of the radon gas continuously monitored using RAD7 instrument for the same period. The dosimeters removed from the calibration chamber and we record the time of removal. The dosimeters films were etched in Na (OH) solution 6.25N for 7h.

Figure 3, shows a plot between radon concentration measurement using RAD7 instrument, and track density on the CR39 detector made the decay of radon and its progenies using standard source of ^{226}Ra. From the best straight-line fitting (R=0.99), we found that the calibration factor K=0.2857±0.01431 Tr cm^{-2} d^{-1} per Bq m^{-3}. Moreover, this measured value in agreement with the value used by Ahmed et al.K= 0.27±0.02 Tr cm^{-2} d^{-1} per Bq m^{-3} [18].

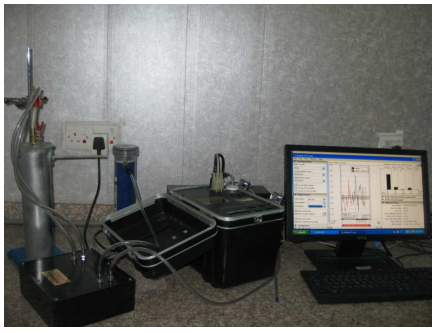

Figure 2: The Calibration Experiment Setup

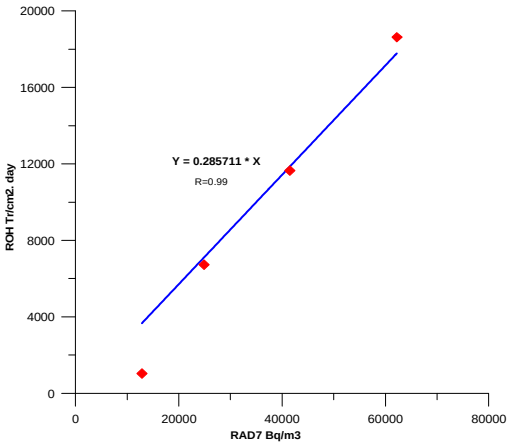

Figure 3: The Calibration Curve, between Track Density and RAD7 Reading

RESULTS AND DISCUSSIONS

Different origin fertilizers have been collected from local positions, and listed in table 1 with their ID. The tracks density made by radon gas on CR39 fixed on top of the chamber for 90 days are listed in the same table. The minimum value for track density is 343±103 Tr/cm^2 made by Potassium sulphate, fertilizer, while the maximum value is 30781±1157 Tr/cm^2 made by Crop Complex, Belgium.

Table 1: The Type of Fertilizer and Their Origin Together with the Masses and Track Density Produced by Irradiate CR39 Track Detector for 90 Days

Sam. No.	Type of Fertilizer and Origin	Mass (kg) Used	Tracks Produced in cm^2
1	NPK, Switzerland	0.190	988∓147
2	Crop Complex, Belgium	0.210	3438∓434
3	NPK, Belgium	0.260	1281∓201
4	Urea-Calacum phosphate, Belgium	0.260	969±129
5	Calacum Nitra, Belgium	0.210	2156±263
6	NPK, Belgium	0.188	1043±121
7	NPK, Belgium	0.215	9406±573
8	Potassium sulphate, Belgium	0.310	343±103
9	Urea, Iran	0.110	781±157
10	Urea, Iraq	0.145	906±169
11	Super phosphate, Iraq	0.200	21468±924
12	NPK –Dap, Jordan	0.214	10438±542
13	Humi-plant, China	0.188	30781±1157
14	NPK, China	0.225	20687±904
15	NPK/SCOTTS, European	0.168	437±117
16	Tobseen-im, Japan	0.90	1044±69

We used equation (1) to calculate radon concentrations in fertilizers ore, according to the measured calibration factor 0.2857±0.01431 Tr cm^{-2} d^{-1} per Bq m^{-3}. The results are listed in table 2 together with the active results of RAD7 instrument. The two types of measurements are correlated as shown in figure 4, where the correlation factor is 87%, means that we have excellent correlation. In other word, we can depend on either one of them, with slight error, to calculate the rest of radon parameters.

Table 2: The Results of Passive (cr39) and Active (rad7) Measurements for the Sixteen Fertilizer Samples

S. N	Passive in Bq/m^3	Active Con. in Bq/m^3
1	38±6	86±10
2	134±18	225±19
3	50±8	47±7
4	38±5	34±7
5	84±11	161±15
6	41±5	44±10
7	366±29	33±4
8	13±4	28±14
9	30±7	29 ±4
10	35±7	99±9
11	835±56	305±19
12	406±30	597±31
13	1197±76	1084±43
14	805±54	218±19
15	17±5	39±9
16	41±6	32±16

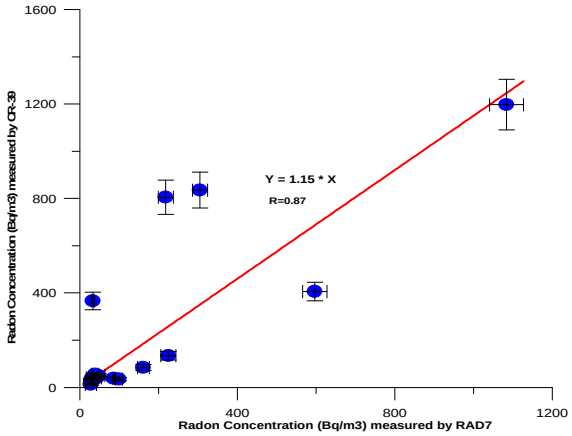

Figure 4: Correlation between the Passive and Active Measurements

The radon exhalation rates, mass exhalation rate and effective radium activity in fertilizers collected from Basrah governorate are given in Table 2, by using equations (2) to (4). It was found that the phosphate samples Hum-plant China has maximum radon concentration 11197±76 Bq/m^3, radon exhalation rate 2.408 Bq/m^2.h, mass exhalation rate 0.0512 Bq/kg. h and effective radium 6.68 Bq/kg. The minimum radon concentration, surface radon exhalation rate, mass exhalation rate and radium effective value is found to be 13±4 Bq/m^3, 26.8 mBq/m^2.h, 0.34 mBq/kg. h and 0.046 Bq/kg respectively in potassium sulphate fertilizer.

Table 3: The Values of the Radon Concentrations, Radon Exhalation Rates and Effective Radium Contents in the Fertilizer Samples

Sample No	Radon Con. Bq/m^3	Area Exhalation Rate mBq/m^2. h	Mass Exhalation mBq/kg. h	Effective Ra in Bq/kg
1	38±6	77.3	1.63	0.215
2	134±18	268.9	5.12	0.678
3	50±8	100	1.54	0.204
4	38±5	75.7	1.17	0.154
5	84±11	168.3	3.21	0.425
6	41±5	81.6	1.74	0.230
7	366±29	735	13.69	1.821

Table 3: Contd.,

8	13±4	26.8	0.34	0.046
9	30±7	61.4	2.22	0.294
10	35±7	70.9	1.95	0.259
11	835±56	16.7	33.58	4.447
12	406±30	816	15.26	2.021
13	1197±76	2408	51.22	6.782
14	805±54	1618	28.76	3.309
15	17±5	112.4	2.68	0.354
16	41±6	81.7	0.36	0.048

The urea fertilizers contained a low detectable source of radon concentration, exhalation rate and effective radium mass. It was also found that one sample of phosphate fertilizers (super phosphate) did show a high detectable radon parameters. It is evident from table 3 that the radon exhalation rate per unit area and unit mass determined with the help of CR-39 detectors of fertilizers are ranging from 16.7 mBq/m^2.h to 2408 mBq/kg. h and 0.34 to 51.22 Bq/kg. h respectively, while the effective radium rate per unit mass determined with the help of the same techniques, ranged from 0.046 to 6.782 Bq/kg. To find out whether these observations are acceptable in comparison with other research works, we found that the work of Saad [23] on phosphate fertilizers introduced a range from 0.020 to 4.125 Bqm^{-2} h^{-1}compared with the range of present work (from 0.027 to 2.4908 Bq $m^{-2}h^{-1}$). The radon exhalation rate per unit mass determined with the help of the same techniques range from 0.003 to 0.658 Bq. kg^{-1} h^{-1}, and our range was; from 0.00034 to 0.051 Bq/kg.h. Finally, one has to mention that radon parameters from fertilizer ores are always higher than fertilizers them self, because the fertilizers are resultantly from mining and the chemical processing of phosphate rocks, which reduces the holes in the material and prevent radon to diffuse out of this host material.

CONCLUSIONS

Samples taken from six types of fertilizers used in Basrah governorate were analyzed for radon concentration and exhalation rates and radium effective mass, using hybrid methods, RAD7 and CR39 alpha detectors. Fertilizers samples type potassium sulphate is found to be much lower than those from NPKs. Thus, the use of potassium sulphate fertilizers fabricated in agriculture is much more appropriate than that from Humi-plant and NPKs. From the present work, it can be concluded that the highest average radon gas concentration in fertilizer samples was found in Humi-plant sample which was 1197±76 Bq/m^3 (China origin), while the lowest average radon gas concentration in fertilizer samples was found in single potassium phosphate sample, which was 13±4 Bq/m^3 (Belgium origin). The present results show that the radon gas concentration in all fertilizer samples is below the allowed limit from (International Commission of Radiation Protection) (ICRP) agency.

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